Tropical Entrainment Time Scales Inferred from Stratospheric N_2O and CH_4 Observations

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Abstract. Simultaneous *in situ* measurements of the long-lived trace gases N_2O and CH_4 were made with a tunable diode laser spectrometer (ALIAS II) aboard the Observations from the Middle Stratosphere (OMS) balloon platform from New Mexico, Alaska, and Brazil during 1996 and 1997. Compact relationships of CH_4 with N_2O are generally seen in the upper troposphere and stratosphere at each latitude. In the extra-tropics, deviations from the normal compact relationship are diagnostic of transport from the tropics or from the polar vortex. Above 26 km, the tropical and extra-tropical relationships differ because transport is slow from the mid-latitude stratosphere to the tropical stratosphere. A simple model is used to calculate the mean time scale for entrainment of mid-latitude air into the tropics. This time scale is estimated to be 7^{+10}_{-7} months for altitudes between the tropical tropopause and 20 km, and 16^{+18}_{-8} months for altitudes between 20 and 28 km. This implies that most of the stratospheric entrainment into the tropics occurs in the lowest portion of the stratosphere.

Introduction

Historically, long-lived tracers have been used to determine the large-scale circulation of the stratosphere because they are conserved during atmospheric motions that are fast compared to chemical production or loss. Brewer [1949] and Dobson [1956] utilized H_2O and column O_3 as passive tracers to infer that air enters the stratosphere at the tropical tropopause, is transported poleward, and returns to the troposphere at middle and high latitudes. More recently, stratospheric CH_4 and N_2O have been studied because of their long photochemical lifetimes ranging from centuries at the tropopause to 1 year in the middle stratosphere. Both gases are produced at the Earth's surface and photochemically removed in the stratosphere. Early measurements of these gases [e.g., Fabian et al., 1981; Jones and Pyle, 1984] showed stratospheric isopleths sloping downward and poleward as expected from the Brewer-Dobson model of stratospheric circulation, but also showed temporal and spatial variations that exceeded instrument precision. Ehhalt et al. [1983] showed that displacements from a mean profile were correlated for CH_4, N_2O, and other long-lived tracers. Therefore, plots of one tracer versus another should remove effects of small-scale, transient air motions.

Two species with photochemical lifetimes much longer than the time scales for quasi-horizontal transport and mixing are in "slope equilibrium," which means the species have the same functional relationship at all latitudes regardless of the geographic distribution of sinks [Plumb and Ko, 1992]. Such species are expected to exhibit a compact correlation (i.e. small variance from a well-defined relationship). However, slow meridional transport between two geographic regions can lead to different tracer *vs.* tracer relationships in each region, as observed in the tropical and extra-tropical lower stratosphere [e.g. Murphy et al., 1993; Volk et al., 1996]. These types of observations led Plumb [1996] to develop the so-called "tropical pipe" model of stratospheric circulation, in which the tropical stratosphere is dynamically isolated from the extra-tropics. Although there is evidence of a dynamical barrier in the subtropics, some material is exchanged between the tropics and extra-tropics. Transport out of the tropics is associated with planetary wave activity, as seen in satellite observations of tracers [e. g., Randel et al., 1993; Waugh, 1993]. There is also evidence from tracer observations that some mid-latitude stratospheric air is transported back into the tropics, but at a considerably slower rate [Avallone and Prather, 1996; Minschwaner et al., 1996; Volk et al., 1996; Schoeberl et al., 1997].

Previous estimates of entrainment rates of mid-latitude air into the tropics utilized tracer data from balloons [Minschwaner et al., 1996], satellites [Minschwaner et al., 1996; Schoeberl et al., 1997], and aircraft [Avallone and Prather, 1996; Minschwaner et al., 1996; Volk et al., 1996]. These studies concluded that the mean entrainment time scale is approximately 12 to 18 months at 20 km and that about 50% of the tropical air at 22 km had been entrained from mid-latitudes. In this paper, we present new, high-accuracy observations of stratospheric N_2O and CH_4 and use them to quantify stratospheric entrainment into the tropics.

Balloon Observations of N_2O and CH_4

The Aircraft Laser Infrared Absorption Spectrometer II (ALIAS II) has measured *in situ* N_2O and CH_4 on six flights of the Observations from the Middle Stratosphere (OMS) balloon platform from New Mexico, Alaska, and Brazil. ALIAS II uses tunable diode lasers to measure N_2O and CH_4 by infrared absorption spectroscopy [Webster et al., 1994]. During balloon ascent and descent, air flows freely through an open-path multipass cell (61.8 m optical path) suspended outside the gondola. Measurements of both gases have an estimated 5% precision and 5% accuracy relative to calibration standards provided by the National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL). The OMS platform has five other instruments, including JPL O_3 [Proffitt et al., 1983], NOAA H_2O [Oltmans et al., 1995], and the NOAA Lightweight Airborne Chromatograph Experiment (LACE) [Elkins et al., 1996]. Simultaneous measurements of pressure by JPL O_3 and temperature (±3 K) by JPL O_3 and NOAA H_2O were used to calculate potential temperature θ (the temperature of an air parcel adiabatically compressed or expanded to 1000 hPa, which is conserved in adiabatic motion). This analysis uses data obtained during both ascent and descent, from the upper troposphere (200 hPa) to the middle stratosphere (9 hPa), excluding the flight of 971111 (yymmdd). Flight dates and locations are summarized in Table 1.

Flight date (yymmdd)	Location	Latitude	Longitude
960610	Ft. Sumner, NM	34° N	104° W
960921	Ft. Sumner, NM	34° N	104° W
970214	Juazeiro do Norte, Ceará, Brazil	7° S	39° W
970630	Fairbanks, AK	65° N	148° W
971111	Juazeiro do Norte, Ceará, Brazil	7° S	39° W
971120	Juazeiro do Norte, Ceará, Brazil	7° S	39° W

Table 1. Flights of the OMS Balloon Payload.

CH_4 and N_2O mixing ratios are highly correlated in the ALIAS II observations (Fig. 1). In the lower stratosphere ($230 \leq N_2O \leq 310$ ppbv), the CH_4 *vs.* N_2O relation is globally compact due to the long photochemical lifetime of each species. However, for N_2O < 230 ppbv, the tropical CH_4 *vs.* N_2O relation deviates from the extra-tropical relation. This indicates that irreversible transport between mid-latitudes and tropics is slower than large-scale mixing within mid-latitudes. A similar separation in CH_4 *vs.* N_2O relationships has been recorded by the Atmospheric Trace Molecule Spectroscopy (ATMOS) instrument aboard the space shuttle [Michelsen et al., 1998], also shown in Fig. 1.

The JPL MkIV interferometer has also measured stratospheric CH_4 and N_2O at mid- and high latitudes [Toon et al., 1989]. Data from two of these flights are superimposed on Fig. 1. In general, the MkIV CH_4 *vs.* N_2O relation is very similar to that measured by ALIAS II, especially for low tracer mixing ratios at high latitudes (blue points). However, for N_2O < 150 ppbv, the mid-latitude MkIV relation (960928) lies between the ATMOS mid-latitude and tropical curves because the air has recently come from the tropics and has not yet mixed completely with the background mid-latitude reservoir. In vertical profile, these data correspond to inversions of both CH_4 and N_2O mixing ratios above 25 km (θ > 630 K), as shown in Fig. 2. The ALIAS II mid-latitude data also have tracer inversions at the same altitude; such inversions are commonly measured above 25 km in the mid-latitude stratosphere [e.g. Kondo et al., 1996] and are manifestations of outflow from the tropics.

In the 970630 Alaska flight, ALIAS II intercepted two narrow layers with anomalously low mixing ratios centered at $\theta \approx 515$ K (20.4 km) and 615 K (24.0 km) (Fig. 2). At these potential temperatures, such low mixing ratios could only be associated with remnants of the Arctic polar vortex. All of the high latitude ALIAS II data that fall below the cluster of mid-latitude data in Fig. 1 are from these layers. These data are similar to the Arctic vortex CH_4 *vs.* N_2O relation of ATMOS because both are mixing lines between the mid-latitude lower stratosphere and descended vortex air. The linear portion of the ATMOS vortex relation corresponds to measurements made below 23 km at 63 to 69°N [Michelsen et al., 1998]. Excluding the vortex remnants, the CH_4 *vs.* N_2O relation measured at 65°N by ALIAS II and MkIV is similar to the mid-latitude relation at 34°N due to rapid meridional transport within the extra-tropics. ATMOS measurements outside the polar vortex confirm this observation [Michelsen et al., 1998]. Although meridional transport is rapid within the extra-tropics, mixing must be relatively slow here to preserve features like the vortex remnants and the mid-latitude tracer inversion.

The measured tropical mixing ratios of N_2O and CH_4 are considerably higher than extra-tropical mixing ratios at a given potential temperature (Fig. 2). This is because tropical air has spent less time in the stratosphere than extra-tropical air, so less photochemistry has occurred. Variability in N_2O and CH_4 is considerably greater for 971120 than 970214, but is correlated with tracers measured by other instruments on the OMS payload. Despite this difference in variability, the mean profiles of N_2O and CH_4 are similar for both flights. In the analysis that follows, it is assumed that a smooth curve through all the tropical data is representative of the tropics.

Tropical Model

The time scale for entrainment of mid-latitude air into the tropics is calculated by comparing the observed tropical mixing ratio profiles to values calculated by a simple tropical model allowing for photochemical loss, vertical advection, and mixing from mid-latitudes. For an air parcel ascending in the tropics with isentropic entrainment of extra-tropical air, the continuity equation is given by [Minschwaner et al., 1996; Volk et al., 1996]:

$$\mathcal{Q}\frac{\partial \chi}{\partial \theta} = -L\chi - \gamma\chi + \left(\frac{\chi_{MID} - \chi}{\tau_{IN}}\right) \quad (1)$$

where $\mathcal{Q}$ is the mass-balanced heating rate (K/day), χ is the tropical mixing ratio (ppbv), χ_{MID} is the mid-latitude mixing ratio at the same potential temperature, $L\chi$ is the local photochemical loss rate (ppbv/yr), $\gamma\chi$ is the long-term growth rate (ppbv/yr), and τ_{IN} is the time scale for entrainment of mid-latitude air into the tropical stratosphere (yr). Mass-balanced heating rates $\mathcal{Q}$ were calculated from a radiative heating model [Rosenlof, 1995], and averaged over latitude (9.5°S - 9.5°N), two years (July 1995 - June 1997), and all longitudes to obtain mean tropical heating rates. Long-term growth rates are included because stratospheric air is older than tropospheric air and thus has a time lag in its initial mixing ratio. These growth rates are global averages of recent tropospheric trends in data collected by the NOAA CMDL global network [Elkins and Dlugokencky, unpubl.].

Photochemical loss includes removal of N_2O by photolysis and reaction with $O(^1D)$, and removal of CH_4 by reaction with OH, $O(^1D)$, and Cl. Twenty four hour average photolysis rates were calculated with a radiative transfer model using observed temperature and O_3 profiles (JPL O_3 measurements from 0 to 30 km, and satellite-based climatology above 30 km) [Salawitch et al., 1994; Minschwaner et al., 1993]. The column abundance of O_3 agrees to within a few DU of measurements from the TOMS satellite instrument. The photochemical model was initialized with measured CH_4, N_2O, and H_2O. NO_y was inferred from the ATMOS relation with N_2O observed in the tropics during November 1994 [M. R. Gunson, pers. comm., 1998]. Cl_y (total inorganic chlorine) and Br_y (total inorganic bromine) were specified based on relations with CFC-11 observed by LACE and also inferred from *in situ* measurements of organic source gases near 20 km [J. W. Elkins, pers. comm., 1998; Wamsley et al., 1998]. Sulfate aerosol surface area was estimated from SAGE II extinction measurements. Twenty four hour average concentrations of OH, $O(^1D)$, and Cl were calculated with this photochemical model [Salawitch et al., 1994] using reaction rates and absorption cross sections from DeMore et al. [1997].

Discussion

In the limit $\tau_{IN} \to \infty$, expression (1) represents vertical advection in the tropics with no mixing in from the extra-tropics, and is solved to obtain unmixed ascent profiles (Fig. 2). Error bars include 30% uncertainty in the N_2O photolysis rate, reaction rate uncertainties from DeMore et al. [1997], 20% uncertainty in radical concentrations, 40% uncertainty in heating rate for $p < 50$ hPa, and 60% uncertainty in heating rate for $50 < p < 90$ hPa (errors added in quadrature). The measured tropical N_2O and CH_4 decrease significantly faster with altitude than the unmixed ascent model, so entrainment of extra-tropical stratospheric air is required to explain the observed mixing ratios [Minschwaner et al., 1996; Volk et al., 1996].

To estimate the time scale for entrainment of mid-latitude air into the tropics, the flight data was binned by potential temperature and expression (1) was solved for τ_{IN} in each bin using polynomial fits to data from ALIAS II and five mid-latitude flights of MkIV. The result is that τ_{IN} increases with altitude but is extremely sensitive to the actual parameters used (the denominator is a small difference between two large numbers). A better approach is to forward-model the tracer profiles by substituting values of τ_{IN} into expression (1) and solving for χ. The simplest solution that reproduces the data well is a short entrainment time for the lower stratosphere and a longer time for the middle stratosphere, as shown in Fig. 3. For N_2O, τ_{IN} is 7^{+10}_{-7} months at altitudes between the tropical tropopause and 20 km (385 K < θ < 450 K), and 16^{+18}_{-8} months for altitudes between 20 and 28 km (450 K < θ < 770 K). For CH_4, τ_{IN} is approximately 8 months between the tropical tropopause and 20 km, increasing to 22^{+38}_{-14} months for altitudes between 20 and 27 km (450 K < θ < 690 K). Given the large differences in lower stratospheric CH_4 between the two tropical flights (Fig. 3), we have greater confidence in the time scales calculated with N_2O. In addition to uncertainties stated above, the error bars of τ_{IN} consider 5% instrument precision and the uncertainty in the observed slope, $\partial\chi/\partial\theta$. At θ = 500 K, these entrainment times are similar to previous results [Minschwaner et al., 1996; Volk et al., 1996]. At θ > 500 K, these times are somewhat longer than the results of Minschwaner et al. [1996], but very similar to the findings of Schoeberl et al. [1997]. The short entrainment time scale below 20 km implies that most of the entrainment into the tropics occurs in the lowest portion of the stratosphere.

In situ observations of N_2O and CH_4 reported here show distinct compact relations for the tropics and extra-tropics. We have used a simple model of the tropical upwelling region to quantify the entrainment rate of mid-latitude air into the tropics. The mean time scale of entrainment increases with altitude from 7^{+10}_{-7} months for altitudes between the tropical tropopause and 20 km to 16^{+18}_{-8} months for altitudes between 20 and 28 km. Two dimensional stratospheric models typically allow for considerably faster exchange of mid-latitude and tropical air than found here. Our results have important implications for calculated trends of mid-latitude ozone [e.g., Volk et al. 1996] as well as the altitude and latitude distribution of NO_y released by future fleets of supersonic aircraft. Once in the tropical stratosphere, NO_y is efficiently lofted to higher altitudes where perturbations to ambient levels of NO_y lead to greater loss of O_3. The entrainment rate profiles shown in Fig. 3 provide important empirical constraints for evaluating multi-dimensional models used to calculate the effects of anthropogenic gases on the global O_3 distribution.

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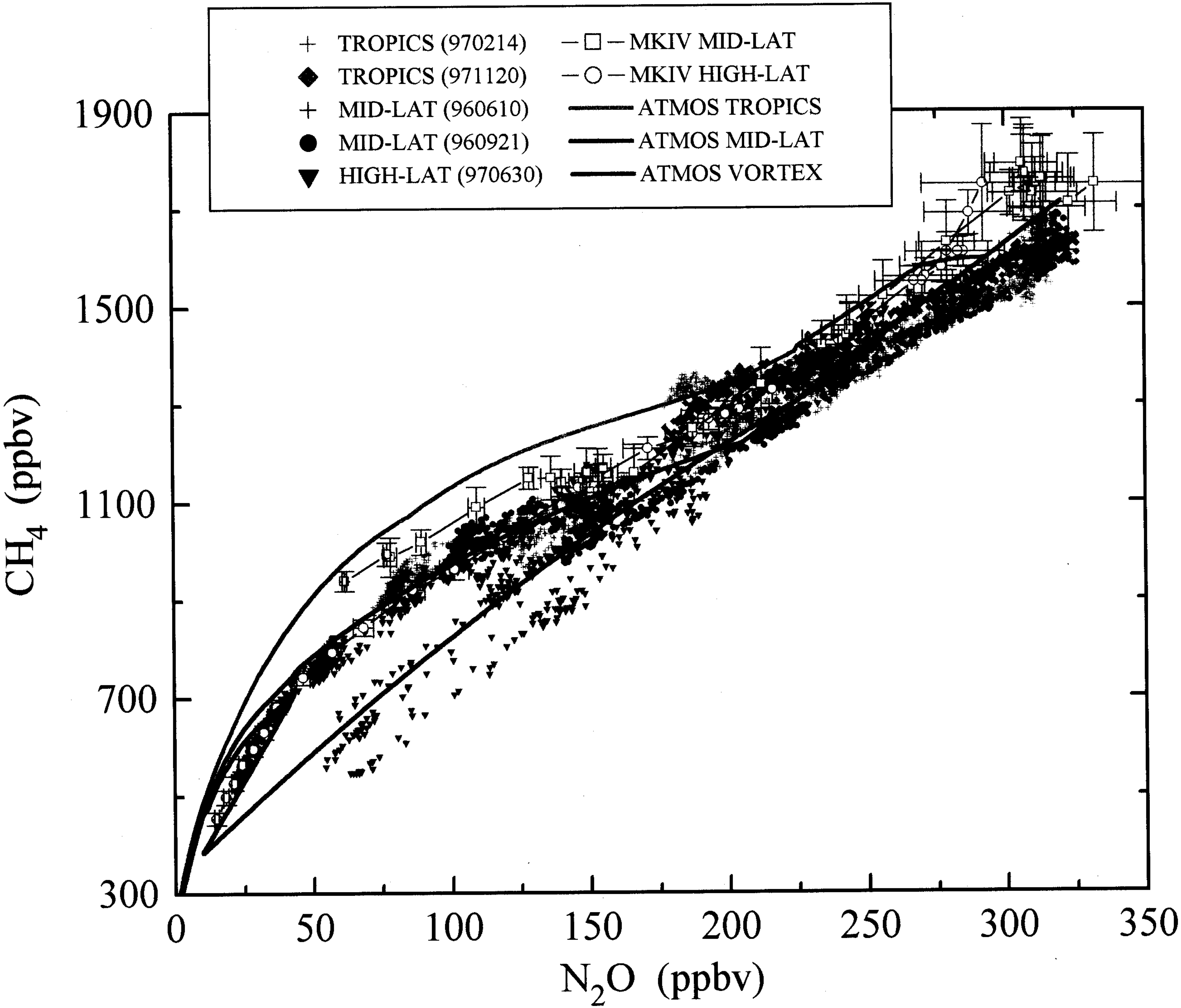
Figure 1. CH_4 versus N_2O volume mixing ratios (red: tropics, green: mid-latitudes, blue: high latitudes). The symbols represent 5 different balloon flights of ALIAS II, including vortex remnants intercepted on 970630. Solid lines are curve fits to ATMOS CH_4 and N_2O data taken aboard the space shuttle [Michelsen et al., 1998]. Lines and symbols with error bars (±1 st. dev.) are MkIV CH_4 and N_2O data from mid-latitudes (960928) and high latitudes (970508).

Figure 2. Vertical profiles of N_2O and CH_4 (red: tropics, green: mid-latitudes, blue: high latitudes). The vertical coordinate is potential temperature θ, which is conserved in adiabatic motion. Both ascent and descent data are shown for ALIAS II (light lines). Lines and symbols with error bars: same as in Fig. 1. The pink line with dashed error bars (±1 st. dev.) is model N_2O and CH_4 in the case of unmixed vertical advection in the tropics. The observed tropical profiles are significantly different than this unmixed ascent model. In addition, the vortex remnants of Fig. 1 correspond to thin layers of low mixing ratios.

Figure 3. Tropical profiles of N_2O and CH_4 as a function of potential temperature θ. The pink line is the same as in Fig. 2. Red symbols are tropical stratospheric measurements of N_2O and CH_4 by ALIAS II. The dark blue line is a third-order polynomial fit to the data. The light blue and green lines are model mixing ratios assuming a constant entrainment time scale, τ_{IN}, as follows: $\tau_{IN}(N_2O)$ = 16 months for 450 K < θ < 770 K and 7 months for 405 K < θ < 450 K; $\tau_{IN}(CH_4)$ = 22 months for 450 K < θ < 690 K and 8 months for 405 K < θ < 450 K.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TROPICS (971120)
MID-LAT (960610)
MID-LAT (960921)
HIGH-LAT (970630)
MKIV MID-LAT
MKIV HIGH-LAT
ATMOS TROPICS
ATMOS MID-LAT
ATMOS VORTEX
1900
1500
1100
700
300
0
50
100
150
200
250
300
350
CH_4 (ppbv)
N_2O (ppbv)

Fig. 1

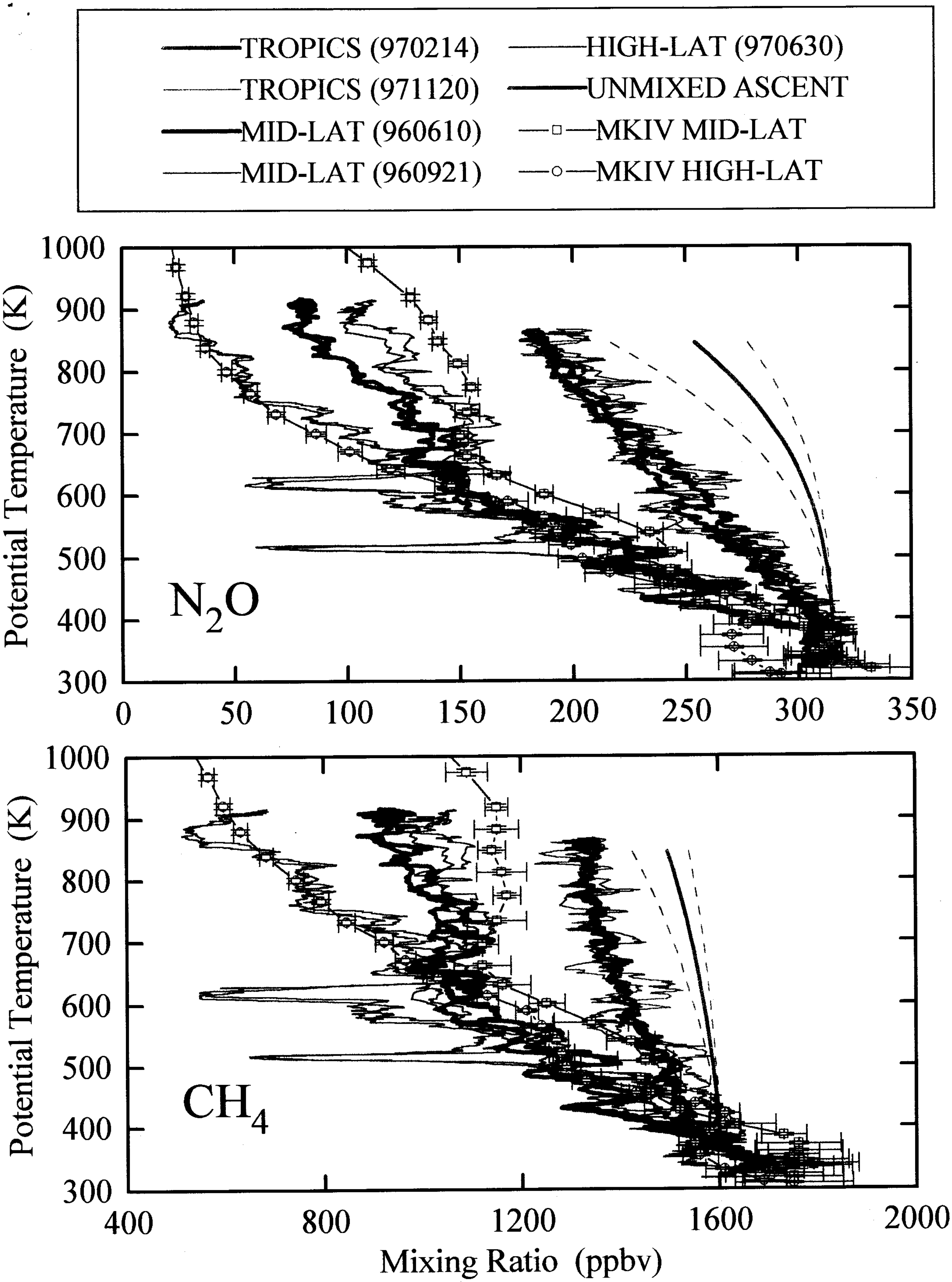
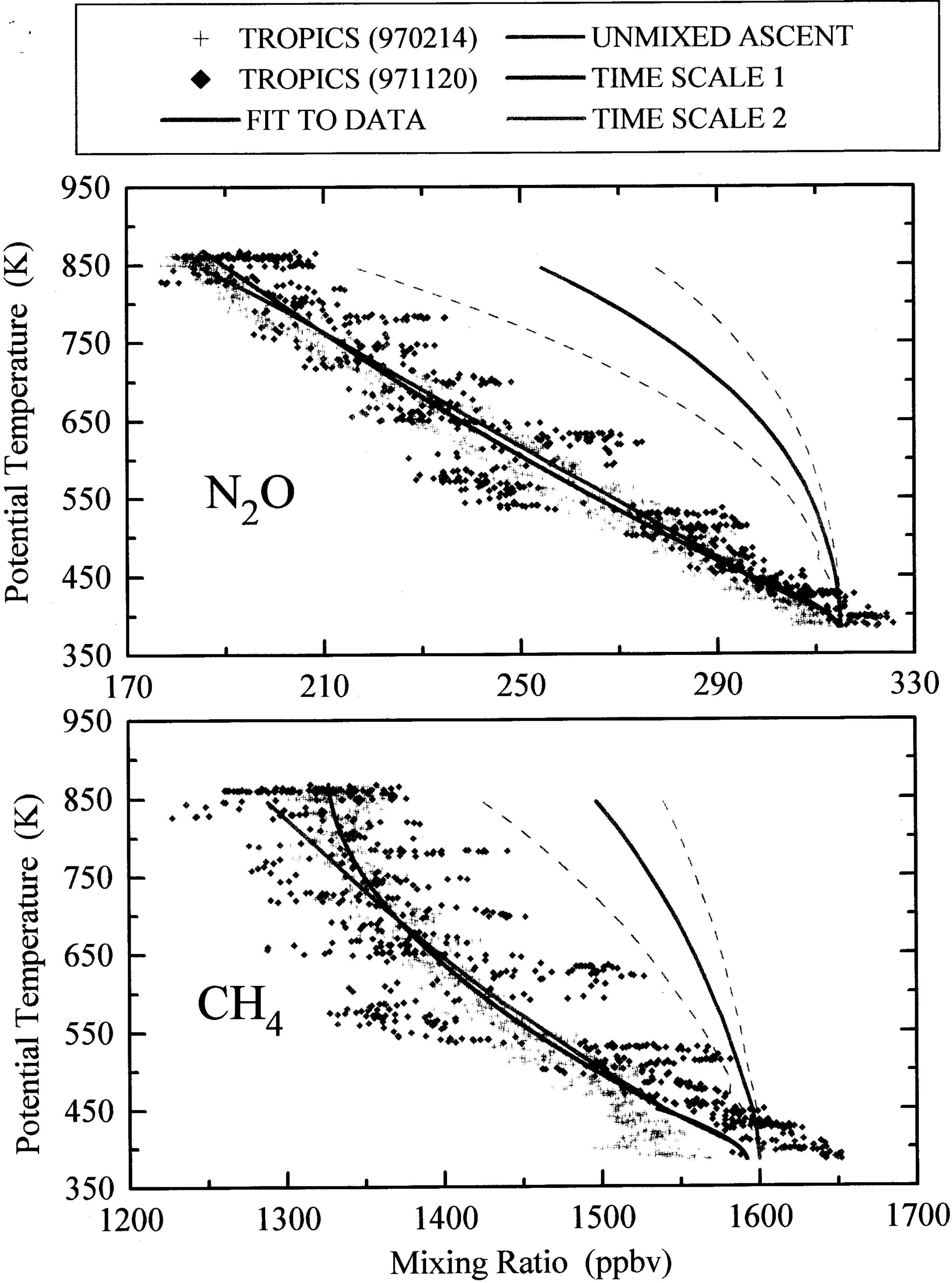

TROPICS (970214)
HIGH-LAT (970630)
TROPICS (971120)
UNMIXED ASCENT
MID-LAT (960610)
MKIV MID-LAT
MID-LAT (960921)
MKIV HIGH-LAT
Potential Temperature (K)
1000
900
800
700
600
500
400
300
N_2O
0
50
100
150
200
250
300
350
CH_4
400
800
1200
1600
2000
Mixing Ratio (ppbv)

Fig. 2



TROPICS (970214)
TROPICS (971120)
FIT TO DATA
UNMIXED ASCENT
TIME SCALE 1
TIME SCALE 2
Potential Temperature (K)
N2O
CH4
Mixing Ratio (ppbv)

Fig. 3